Kinetics and Structures of Self-assembled Nanocrystal Superlattices

Scientific Achievement

Self-assembly of nanocrystals has been considered as one of the promising candidates for making future electronic and optical devices. However, even for the simplest case of drying a nanocrystal colloidal droplet, a variety of self-assembled structures can be formed. Presumably, this is due to different experimental conditions, such as particle sizes, ligand chain length, solvent polarity, evaporation rate, etc. On the other hand, the physical properties of different assemblies can vary dramatically because of the structural difference. Therefore, understanding how self-assembly depend on the experimental condition is crucial to control the self-assembling process. We have adopted *in situ* small angle x-ray scattering and optical microscopy experiments to study the dynamics of the self-assembly under various experimental conditions. (Bigioni et al. *Nature Materials*, in press; Narayanan et al. *Phys. Rev. Lett.* **2004**, 93, 135503). We have shown evaporation kinetics and particle interactions with the liquid-air interface are two key parameters to tune the structure of nanocrystal superlattices. Assembly of nanocrystals into 2D superlattices occurs preferentially at the liquid-air interface under the fast initial evaporation condition and a strong particle interaction with the liquid-air interface. However, a slow evaporation kinetics and weak particle liquid-air interface interaction leads to 3D superlattices that show preferential structural orientations on the substrate (Li et al. manuscript in preparation).

Significance

Our work is the first *in situ* experiment of this kind designed to study the dynamics of nanocrystal self-assembling process. It also opens up a variety of new venues to control the self-assembly process on the nanoscale. Therefore, it will have a significant impact on bottom up self-assembly using nanometer scale building blocks in general. This also opens up a new horizon for the fabrication of technologically important nanocrystal thin film materials for sensors, optical devices and magnetic storage media.

Performers

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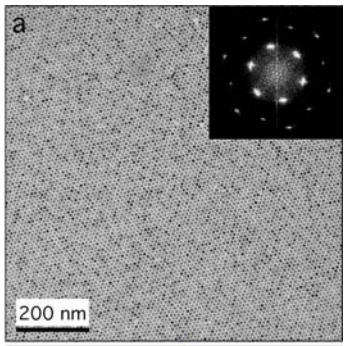
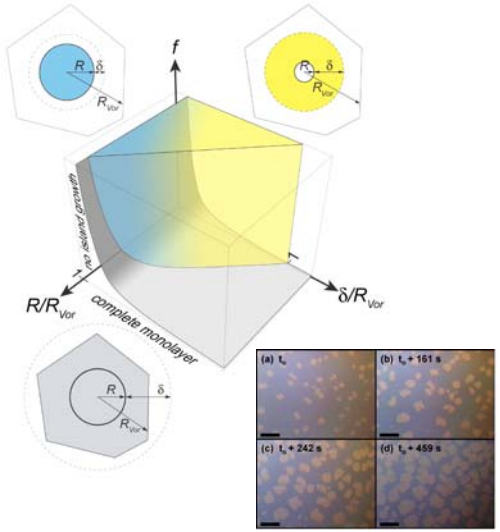
Motivation and Scientific Impact: Colloidal droplet evaporation is typically a far-from-equilibrium process that is affected by fluid flows and solvent fluctuations. Our recent experiments, however, found that controlling this evaporation process can lead to highly-ordered 2D and 3D Au nanocrystal superlattices. Through *in situ* small angle x-ray scattering and optical microscopy, we show that evaporation kinetics and particle interactions with the liquid-air interface are two key parameters to tune the structure of superlattices. This opens up new horizons for the fabrication of technologically important nanocrystal thin film materials for sensors, optical devices and magnetic storage media.

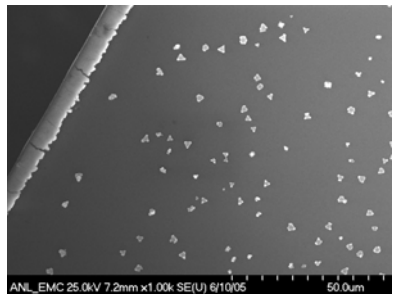
2D

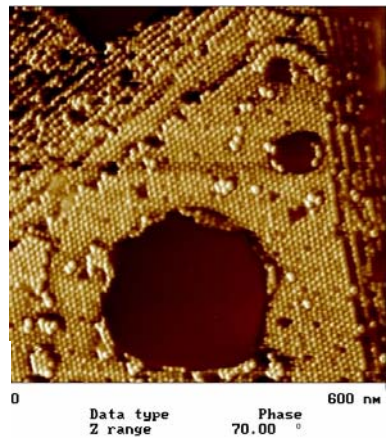


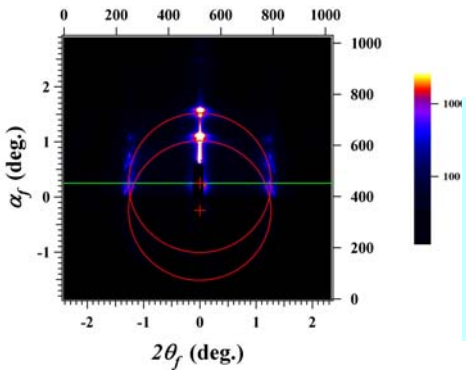


- Self-assembly is controlled by evaporation kinetics and particle interactions with the liquid-air interface.
- 2D nanocrystal superlattices form at the liquid-air interface.
- 2D domains grow either in exponential, linear and intermediate growth laws.

3D







- 3D nanocrystal superlattices form near the substrate.
- 3D domains show preferential orientation on the substrate.
- The lattice spacing decrease at elevated temperature before sintering.

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